

LAGRANGIAN TRANSPORT CALCULATIONS USING  
UARS D/FJ A. PART II: OZONE

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## Abstract

Trajectory calculations are used to examine ozone transport in the polar winter stratosphere during periods of UARS observations. The utility of these trajectory calculations for determining mass transport has been verified previously using UARS observations of long-lived tracers. In the middle stratosphere, the ozone behavior observed by MILES in the polar vortex is reproduced by this purely dynamical model. Calculations show the evolution of ozone in the lower stratosphere during early winter to be dominated by dynamics in Dec 1992 in the Arctic, but the calculations suggest the possibility of some discernable chemical loss in Jun 1992 in the Antarctic. Calculations confirm that chemical destruction of ozone was important in late winter during Aug/Sep 1992 in the Antarctic and during Feb/Mar 1993 in the Arctic. Estimates of differences between the calculated and observed fields suggest that dynamical changes are unimportant in estimating the amount of chemical loss in the Antarctic. In the Arctic, during Feb/Mar 1993, estimates suggest that vortex averaged chemical ozone destruction may have been as much as  $\pm 1\%$ /day near the 465 K isentrope in the lower stratosphere.

## 1. Introduction

An important and complex problem in understanding the distribution and evolution of ozone in the stratosphere is that of distinguishing chemical from dynamical effects. One approach that can be used where observations of ozone and meteorological fields are available is to model ozone transport using winds derived from observations, but ignoring chemical processes. A comparison of fields thus modeled with observed fields indicates which features of the observed fields may be explained by dynamics.

Manney et al. (1994a, hereafter "Part I") study the transport of passive tracers observed by the Cryogen Limb Array Lalon Spectrometer (CLALS) and Microwave Limb Sounder (MLS) instruments on the Upper Atmosphere Research Satellite (UARS). They use trajectory calculations as a kind of transport code, by filling the stratosphere with air parcels, and associating with each parcel the observed value of a tracer mixing ratio on the initial day. Four time periods are studied: 3 Dec 1992 through 2 Jan 1993 and 4 Feb through 16 Mar 1993 in the northern hemisphere (NH); 15 Jun through 5 Jul 1992 and 18 Aug through 7 Sep 1992 in the southern hemisphere (SH).

Waters et al. (1993a,b) show that, during the two SH time periods mentioned above, elevated values of chlorine monoxide (ClO), the dominant form of reactive chlorine that destroys ozone in the daytime lower stratosphere, were present in most of the sunlit portion of the polar vortex in the lower stratosphere. Slightly enhanced ClO was observed at some times in the NH in Dec 1992 (Manney et al. 1994b). In Feb 1993, enhanced ClO values in the NH were comparable to those in the SH in August 1992 (Waters et al. 1993a). Manney et al. (1994b) show a significant decrease in ozone in the polar lower stratosphere during Feb and early Mar 1993; by comparing observations of ozone with expectations based on the evolution of passive tracer measurements, they argue that this decrease cannot be explained solely by dynamics. Together with the observations of high ClO, this demonstrates that chemical loss occurred. However, they do not quantify the amount or spatial extent of chemical loss, except by comparison to the Antarctic ozone hole and to the ozone distribution in the Arctic in 1978-1979, a year in which chemical depletion in the lower stratosphere was not expected.

In the following, we examine ozone transport using the same trajectory runs shown in Part I. Since these runs successfully reproduced the observed distributions of  $N_2O$  and  $CH_4$ , we have some confidence that the trajectory scheme and the wind fields it employs would reproduce the general features of the observed extratropical ozone distribution in the middle and lower stratosphere, if ozone were nearly conserved over the thirty-day

time periods considered here. Thus, differences between the calculated and observed ozone fields are indicative of chemical loss. By examining the transported ozone in the lower stratosphere, and comparing to the transport of passive tracers shown in Part I, and to observed ozone, we place some limits on how much of the ozone evolution can be explained solely by transport processes.

## 2, Data and Analysis

The trajectory calculations are described in Part I and in Manney et al. (1994c). Horizontal winds are from the United Kingdom Meteorological Office (UKMO) data assimilation system (Swinbank and O'Neill 1993) and vertical velocities from a recent version of the middle atmosphere radiation code MIDRAD, an earlier version of which is described by Shine (1987). Temperatures used in the radiation code are from the UKMO data; the radiation code uses MLS ozone, except for the Jun 1992 run when continuous reliable ozone data were not available. Manney et al. (1994c) discuss some limitations of the radiation calculation, and differences seen between heating rates calculated using climatological and MLS ozone. The trajectory code (Manney et al. 1994c) uses a standard fourth-order Runge-Kutta scheme. Winds and temperatures are interpolated linearly in time, from the once daily values to the trajectory time step (1/2 hour). Heating rates are recalculated every 3 hours using interpolated temperatures, and are interpolated linearly to the trajectory time step between calculations. Further details of the trajectory calculation are given by Manney et al. (1994c), and of the initialization of parcels in Part I.

The Rossby-Ertel potential vorticity (P<sub>V</sub>) is also calculated from the UKMO data (Manney and Zurek 1993) and is compared with calculated and observed tracer fields. When examining vertical cross-sections, P<sub>V</sub> is scaled in "vorticity units" using a standard atmosphere value of the static stability (Dunkerton and Delisi 1986, Manney and Zurek 1993); this gives a similar range of values for P<sub>V</sub> on isentropic surfaces throughout the

stratosphere.

The ozone data are from the M1S 205 GHz radiometer; they have horizontal resolution of  $\approx 400$  km and vertical resolution of  $\approx 4$  km. The UARS M1S instrument is described by Barath et al. (1993), and retrieval methods are summarized by Waters et al. (1993a). Precisions (rms) of individual ozone measurements for the altitudes examined here are  $\approx 0.3$  ppmv, with absolute accuracies of  $\approx 5\text{--}15\%$  (UARS data validation report, in preparation). Ozone data are gridded using Fourier transform techniques that separate time and longitude variations (Elson and Froidevaux 1993).

Air parcels are initialized on the grid described in Part 1, on 15 June 1992 and 18 Aug 1992 in the **S11**, and 3 Dec 1992 and 14 Feb 1993 in the **N11**; the trajectory code was run for 20 days for the June 1992 case, and 30 days for each of the other cases. The general motions of air parcels during these time periods were described by Manney et al. (1994c).

The method used to grid the calculated fields from values at the advected parcel positions is described in Part I. Results are also examined by interpolating observations to the advected parcel positions; this is a straightforward linear interpolation in three dimensions.

Ozone is mostly produced in the mid-stratosphere at low latitudes (e.g., Brasseur and Solomon 1984). The purely advective calculation described in Part I will not maintain that source over the time period of the runs considered here; test calculations for Feb and Mar 1993 showed that the calculated fields very quickly diverged from the observed fields in low latitudes because there was no mechanism for maintaining high ozone mixing ratios at the equator. To alleviate this problem, while still maintaining dynamically controlled fields in middle and high latitudes, we parameterize standard gas phase ozone photochemistry by relaxing the ozone field to values observed by M1S each day during the gridding procedure, as described by Sabutis (1994). The relaxation time constant is dependent on latitude, altitude, and day of year, and is based on Figure 11 of Garcia and

Solomon (1985). Below  $\approx 30$  km, and in middle and high latitude regions for all altitudes considered here, relaxation time constants are much longer than the 30 days for which these runs are done. Hence, the net effect of this procedure is to force parcels that are in low latitudes in the middle and upper stratosphere on a given day to high ozone values. Comparisons between fields gridded using this procedure, and the purely advective one described in Part 1 showed differences at latitudes greater than  $\approx 30^\circ$  in the middle stratosphere only when equatorial air is being drawn into high latitudes. The procedure, with the time constants used here, results in holding ozone close to its observed values at latitudes equatorward of  $\approx 30$ -- $40^\circ$  at altitudes above  $\approx 30$  km, and allowing advection to determine its distribution elsewhere. This is acceptable for this study which focuses on middle and high latitudes in the middle and lower stratosphere.

### 3. Results

As in Part I, we compare the average characteristics of the fields with observations interpolated to the parcel positions. Figure 1 shows for each period, at 465 and 840 K, the average, minimum, and maximum values of observed ozone mixing ratios interpolated to the parcel positions, for those parcels that were initially inside the vortex, as defined by a PV contour in the region of strong PV gradients. As described in Part 1, any changes in these from the first day represent discrepancies between the observed field and the field that would be calculated by pure advection.

It is expected that at 840 K in the polar winter regions, the behavior of ozone will be controlled by transport (e.g., Hartmann and Garcia 1979). This is verified in our calculations, in that the plots of parcels initialized at 840 K show no obvious trends. There is a slight upward trend in the last  $\approx 10$  days of the run started 14 Feb 1993, similar to that shown in Part 1 for  $N_2O$  and  $Cl_2$ . Since 840 K is near (but below) the ozone maximum, and vertical gradients are weak, this is further evidence that this increase is due mainly to

the horizontal evolution of the fields, as suggested in Part I.

No apparent trend is seen in the 465 K ensemble average ozone mixing ratios in the two early winter time periods, suggesting that dynamics is still the main factor in the evolution of these fields. Minimum values at 465 K in Dec 1992 show a steady downward trend, suggesting the possibility of some localized non-dynamical changes that are not widespread enough to affect the average. In both of the late winter cases, the observed ozone at the parcel positions decreases steadily. Since the corresponding plots for passive tracers shown in Part I do not show a similar trend, we conclude that processes other than dynamics are important; this is expected in the SH late winter, and is consistent with the findings of Manney et al. (1994b) for the NI 1 late winter of 1993,

Figure 2 shows synoptic maps from the gridded calculated fields compared with observations at 840 K, for the NI 1 and SH during late winter periods. As was the case in Part I with the passive tracer fields, the transport calculation does a good job of reproducing the extent and shape of the region of low ozone in the polar vortex, and the tongues of high ozone that are drawn up around the edge of the vortex. On 2. and 10 Mar 1993, observations show a region of low ozone in the anticyclone where the transport calculations suggest there should be high values. Detailed trajectory calculations by Manney et al. (1994, paper in preparation) indicate that this feature cannot be generated by transport processes alone.

For an overview of the ozone evolution in the mid-stratospheric polar vortex, Figure 3 shows vortex averaged calculated and observed ozone for each of the four time periods, from 655- 1300 K. The trajectory calculations reproduce most features of the observed (distributions below  $\approx 1000$  K, indicating that most of these variations are due to dynamical processes. The differences between observations and calculations for Dec 1992 suggest that the radiation code used produces diabatic descent that is too strong near 840 K, consistent with our finding from the study of passive tracers in Part I. Some irregularities are seen in the calculated fields at the highest levels toward the end of the runs. This is

due to the fact that by this time, there are few parcels left at those levels in the polar regions (Part I).

Figure 4 shows synoptic maps at 465 K of calculated and observed ozone throughout the late winter time periods. We note that tongues of high ozone are frequently stripped off the edge of the vortex in the NH, and occasionally in the SH. The trajectory calculation reproduces these features at least through  $\approx$  day 20 of the run. Within the vortex, the calculated fields in Fig. 4 show how the 465 K ozone field should look if no chemical destruction of ozone occurred after the initial day of the simulation. In the NH during Feb/Mar 1993, transport causes ozone to increase gradually throughout the period, while observations show a steady decrease. The predicted increase in the NH due to dynamical processes is consistent with the behavior inferred from passive tracer observations and past studies of earlier years (Manney et al. 1994b, Part 1). In the Antarctic, ozone mixing ratios predicted by transport are nearly constant, while observed values decrease rapidly. The early winter fields at 465 K (not shown) show no appreciable differences between observed and calculated fields.

Figure 5 shows vortex averages of calculated and observed ozone in the lower stratosphere (420 - 655 K) for each of the four time periods. Diabatic descent rates estimated from the calculated fields as described in Part I are consistent with those obtained from calculations based on CLAES passive tracer fields, except during Aug/Sep 1992, when the value obtained using calculated ozone is  $\approx 0.4$  K/d ( $d\theta/dt$ ). This is consistent with the values estimated by Manney et al. (1994c) using the parcel positions directly, and thus supports the view, stated in Part 1, that there may be problems with the CLAES fields in the lower stratosphere on the initial day of this run, which were used for the estimate in Part I.

The observed and calculated vortex averages in Dec 1992 are consistent with three obtained from passive tracer fields, and thus suggest that the evolution of ozone in the lower stratosphere during this time period is dominated by dynamical processes. In Jun

1992, observed vortex averaged ozone shows a gradual decrease between  $\approx 500$  and 600 K, starting about the middle of the time period ( $\approx 22$  Jun). This decrease is not seen in the calculated ozone, nor is it consistent with the behavior of passive tracers shown in Part 1. At  $\approx 550$  K, if changes in the ozone field were caused by vertical motion, the observed evolution would imply a vortex averaged diabatic *ascent* of  $\approx 0.7$  K/d around that level. Since Manney et al. (1994c) and Part 1 conclude that there is in fact quite strong diabatic *descent* during this period (between  $\approx 0.8$  and  $\approx 1.3$  K/d), and since Manney et al. (1994c) show that there is very little mixing across the PV contour used here to define the vortex average, it is unlikely that the observed changes in ozone between 500 and 600 K during Jun 1992 could be caused by dynamics alone. Waters et al. (1993b) showed significant enhancement of ClO at 22 hPa (near 600 K) at this time, thus chemical loss would be expected. However, the changes shown here are quite small.

For the two late winter cases, it has previously been shown that depletion of ozone by chlorine chemistry is important (Waters et al. 1993a,b; Manney et al. 1994b). The trajectory calculations confirm that if only dynamical processes were important, ozone should increase at levels throughout the lower stratosphere. Using estimates given in Part 1 for diabatic descent rates, at the end of the Feb/Mar 1993 run, dynamics would be expected to result in  $\approx 3.3$  to  $3.5$  ppmv vortex averaged ozone at  $\approx 470$  K, as opposed to the observed value of  $\approx 2.7$  to  $2.9$  ppmv. In Aug/Sep 1992 in the SH, our estimates suggest that dynamical processes would result in  $\approx 3.0$  to  $3.2$  ppmv vortex averaged ozone, as opposed to the observed value of  $\approx 1.9$  to  $2.0$  ppmv.

Figure 6 compares, as a function of PV and  $\phi'$ , differences in calculated and observed ozone in the lower stratosphere between the first and last days of the two S} I runs and the Feb/Mar 1993 NH run, Parts (c) and (e), from observations are similar to figures shown by Manney et al. (1994b), but for a shorter time interval, so changes are generally somewhat smaller. The predicted fields show, over most of the domain, an increase in ozone, due to the diabatic descent of higher ozone from above. In Jun 1992,

observed changes show a decrease in ozone in the polar vortex between 500 and 600 K; while this is not inconsistent with chemical loss occurring at this time, other observed decreases, such as the widespread decrease above 750 K, are difficult to explain.

Comparison of the observed and calculated fields for Aug/Sep 1992 suggests that changes above  $\approx 750$  K throughout the PV domain shown are explainable by dynamics alone. Changes below this level are dominated by chemistry. In the NH 1 in Feb/Mar 1993, changes above  $\approx 700$  K are generally reproduced with the dynamical model. Although the calculated values suggest a small decrease in ozone between  $\approx 550$  and 650 K, and  $\approx 1.4$  and  $2.0 \times 10^{-4}$  s<sup>-1</sup> in PV, this decrease could explain only a small fraction of observed changes,

Figure 7 shows, for the SH early winter time period, an estimate of the non-dynamical changes in ozone (presumed to be chemical destruction) after the initial day of the runs in the vortex average, determined by subtracting the observed ozone from the calculated value at 585 K on each day. The calculated non-dynamical change is compared to the observed change from the value on the first day. In the SH early winter this suggests that actual chemical loss might be  $\approx 0.1$  ppmv more than the observed ozone decrease. If, as suggested by the passive tracer study (Part 1) the trajectory calculations underestimate the diabatic descent in the SH, the actual chemical loss could be somewhat larger. However, the ozone decrease seen here is small ( $\approx 0.15$  to  $0.25$  ppmv), probably within the uncertainties in the data and analyses.

Figure 8 shows similar plots for the NH and S 1 late winter time periods. In the NH in Feb/Mar 1993, the calculations suggest that the loss due to non-dynamical processes (chemistry) may have been  $\approx 0.25$  ppmv greater than the observed decrease over this 30 day period, as the chemical loss is superimposed on a dynamically produced increase in ozone. This would raise the estimate of chemical ozone depletion at 465 K during this time period from the observed change of  $\approx 0.7$  %/day (Manney et al. 1994b) to  $\approx 1$  %/day. Since Part 1 showed that the trajectory calculation tended to overestimate the diabatic

descent here, we regard this as a upper limit on the actual chemical loss.

The two curves for the Aug/Sep 1992 time period are nearly indistinguishable. The trajectory calculation did tend to underestimate the descent here, but even the largest estimate suggested in Part 1 by either observations or calculations would result in an additional estimated chemical loss of only  $\approx 0.1$  ppmv, a small percentage of the total observed loss at this time. The amount of ozone decrease in the vortex is much greater in the S1-1 than in the NH. in addition, the SH dynamical situation is much more quiescent, so there is less descent of higher ozone from above. Thus, extracting the effects of dynamics does not have an appreciable effect on the estimate of overall chemical ozone loss in the SH vortex. in contrast, diabatic descent is larger in the NH, and net changes in ozone are smaller, so dynamical effects must be accounted for to reliably estimate the amount of chemical ozone destruction.

#### 4. Conclusions

Trajectory calculations are used to examine ozone changes in the polar winter stratosphere for a period of UARS observations. The utility of these trajectory calculations for determining mass transport was verified in Part I. We focus on comparisons with observations which provide information on where and when the observed evolution of ozone can or cannot be explained purely by dynamical processes. Four time periods were considered, covering early and late winter in the NH and the SH, when UARS MLS ozone observations were available for high latitudes.

in the middle stratosphere, ozone in the polar vortex is shown to behave in a manner consistent with the behavior of passive tracers discussed in Part 1. There are exceptions to this dynamical consistency in mid-latitudes; one instance of this is shown here.

In the lower stratosphere, during Dec 1992 in the NH, the average behavior of ozone was again consistent with that of passive tracers (Part 1), although there is a suggestion of

some local non-dynamical changes. Observations showed some enhancement of ClO in the lower stratosphere at this time (Manney et al. 1994b); our calculations suggest that there was not enough chemical loss to be detectable over the dynamically caused changes. In the SH early winter, in Jun 1992, considerably more ClO enhancement was seen (Waters et al. 1993b), and the calculations presented here suggest the possibility of some discernable chemical loss.

It has been previously shown that chemical destruction of ozone was important during the Aug/Sep 1992 SH period (Waters et al. 1993b, Manney et al 1993), and during the Feb/Mar 1993 NH period (Manney et al. 1994b). Our calculations confirm these results. Estimates of the differences between the calculated and observed fields suggest that the dynamical changes do not significantly affect estimates of the amount of chemical loss in the SH. in contrast, in the NH during Feb/Mar 1993, the estimates suggest that vortex averaged chemical destruction of ozone at 465 K in the lower stratosphere may have been as much as  $\approx 1\%$ /day, compared with the  $\approx 0.7\%$ /day observed decrease in ozone.

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## Figure Captions

Figure 1. Average (circles), minimum (triangles) and maximum (squares) observed  $O_3$  mixing ratios (ppmv) for the ensemble of parcels that were initially inside (i.e., at higher PV than) the  $1.4 \times 10^{-4} \text{ s}^{-1}$  scaled PV contour (-PV in the SH). Observed values are interpolated to parcel positions on each day, and the average, minimum and maximum of these values are shown. Values are shown for parcels started at 465 and 840 K in each of the four periods.

Figure 2. Synoptic maps of  $O_3$  at 840 K from observations and trajectory calculations, on days 0, 8, 16 and 24 of the late winter runs in the NH and SH. The projection is orthographic, with  $0^\circ$  longitude at the bottom of the plot in the NH and at the top in the SH and  $90^\circ\text{E}$  to the right; dashed lines show  $30^\circ$  and  $60^\circ$  latitude circles. Several PV contours in the region of strong PV gradients are overlaid on the plots of observed values.

Figure 3. Time series of vortex-averaged  $O_3$  mixing ratios (ppmv) as a function of  $\theta$  in the mid-stratosphere (655 K-- 1300 K). Vortex-averages are area weighted, and are computed using the  $1.4 \times 10^{-4} \text{ s}^{-1}$  scaled PV contour (-PV in the SH) as a approximate definition of the vortex edge. Values between 5.2 and 5.4 ppmv are shaded,

Figure 4. Synoptic maps of ozone mixing ratios (ppmv) at 465 K from observations and trajectory calculations, on days 0, 4, 8, 12, 16, 20, 24, and 28 of the run started 14 Feb 1993 in the NH, and on days 0, 8, 16, and 24 of the run started 18 Aug 1992 in the SH. The layout is as described for Fig. 2.

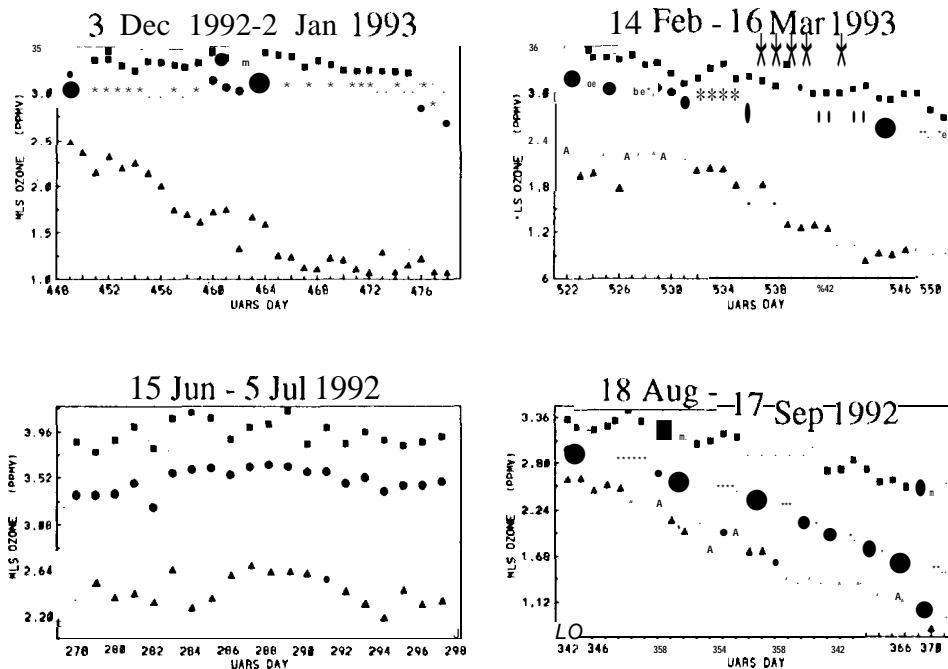
Figure 5. As in Figure 3, but for the lower stratosphere (420 K--655 K). "The scaled PV contour used is  $1.2 \times 10^{-4} \text{ s}^{-1}$  (-PV in the SH), and mixing ratios between 2.0 and 2.2 ppmv are shaded.

Figure 6. Difference plots of ozone mixing ratios as a function of PV and  $\theta$  in the lower stratosphere, from the trajectory calculations and the observations, for day 20 minus day 0 of the run started in Jun 1992 in the SH, and for day 29 minus day 0 of the runs started in Aug 1992 in the SH and Feb 1993 in the NH, The polar vortex is at PV values  $> \approx 1.0-1.4 \times 10^{-4} \text{ s}^{-1}$ , -PV is used in the SH.

Figure' 7, Estimated "non-dynamical" ozone change between 15 Jun and 5 Jul 1992. Solid line shows the difference between the observed and calculated vortex-averaged ozone mixing ratio on each day. Dashed line shows the observed change in vortex-averaged mixing ratio from 15 Jun.

Figure 8. As in Fig. 7, but for (a) 14 Feb through 15 Mar 1993 in the NH, and (b) 18 Aug through 16 Sep 1992 in the SH.

# PARCELS STARTED AT 465 K, PV > 1.4



# PARCELS STARTED AT 840 K, PV > 1.4

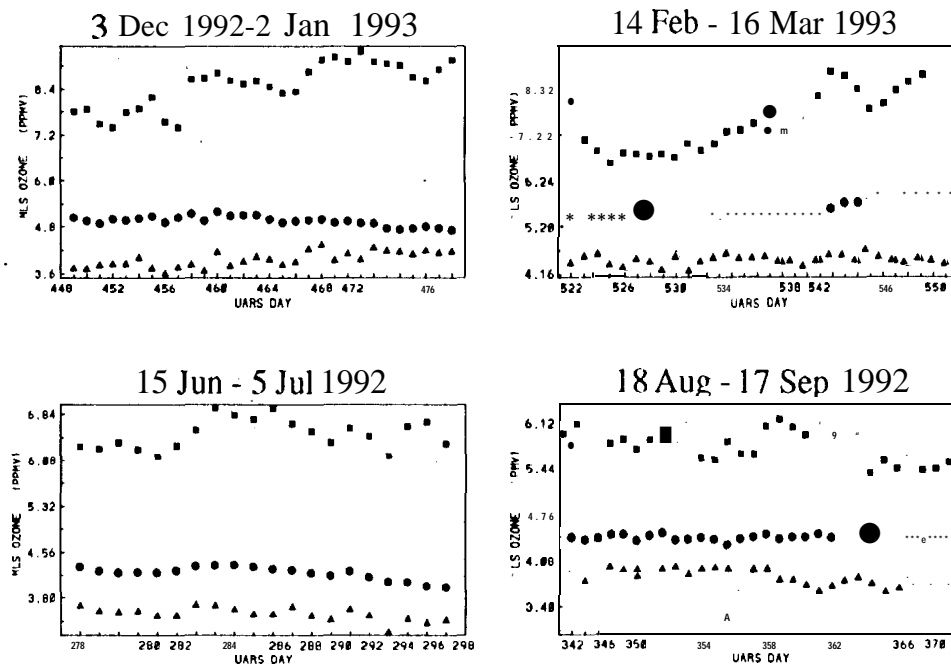
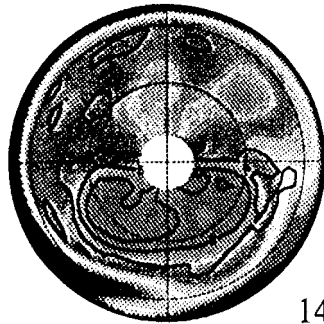


Fig. 1

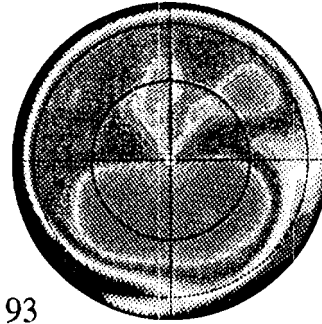
840 K Ozone (ppmv)

OBS

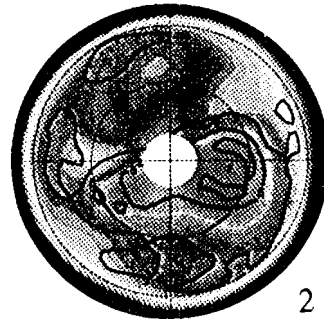
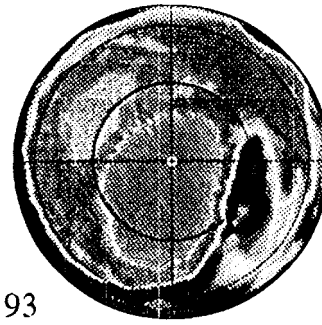
TRAJ



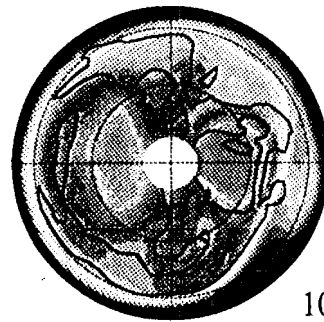
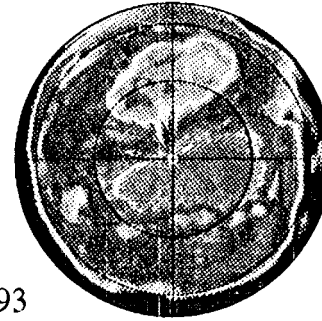
14 Feb 93



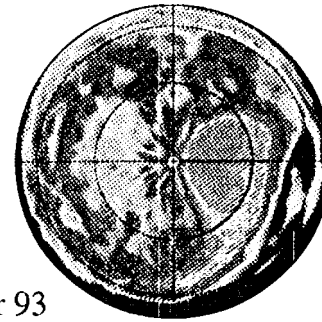
22 Feb 93



2 Mar 93



10 Mar 93

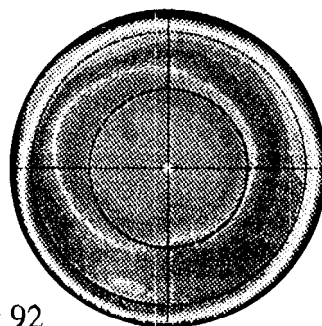
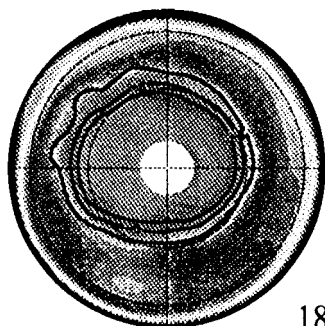


*Fig. 2a*

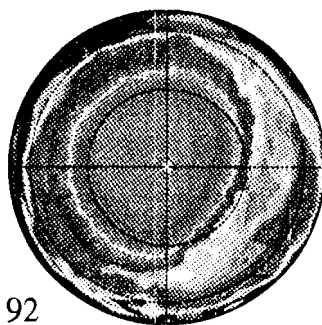
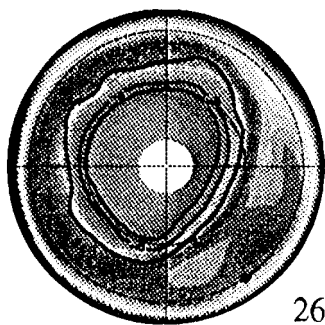
840 K Ozone (ppmv)

OBS

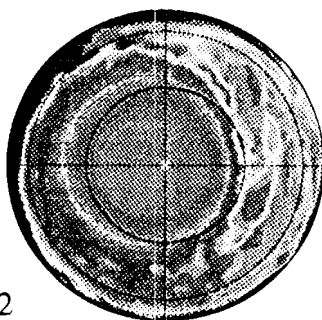
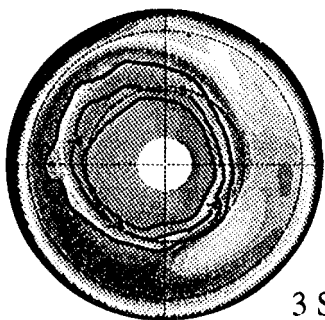
TRAJ



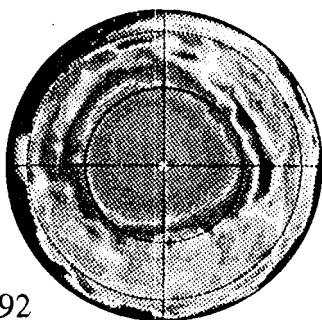
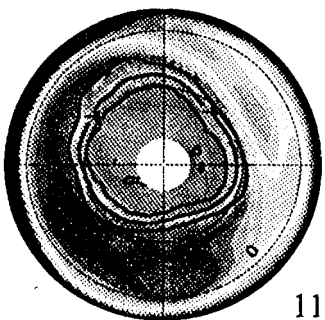
18 Aug 92



26 Aug 92



3 Sep 92



11 Sep 92



*Fig. 2 b*

# VORTEX-AVERAGED OZONE (PPMV) MIDDLE STRATOSPHERE

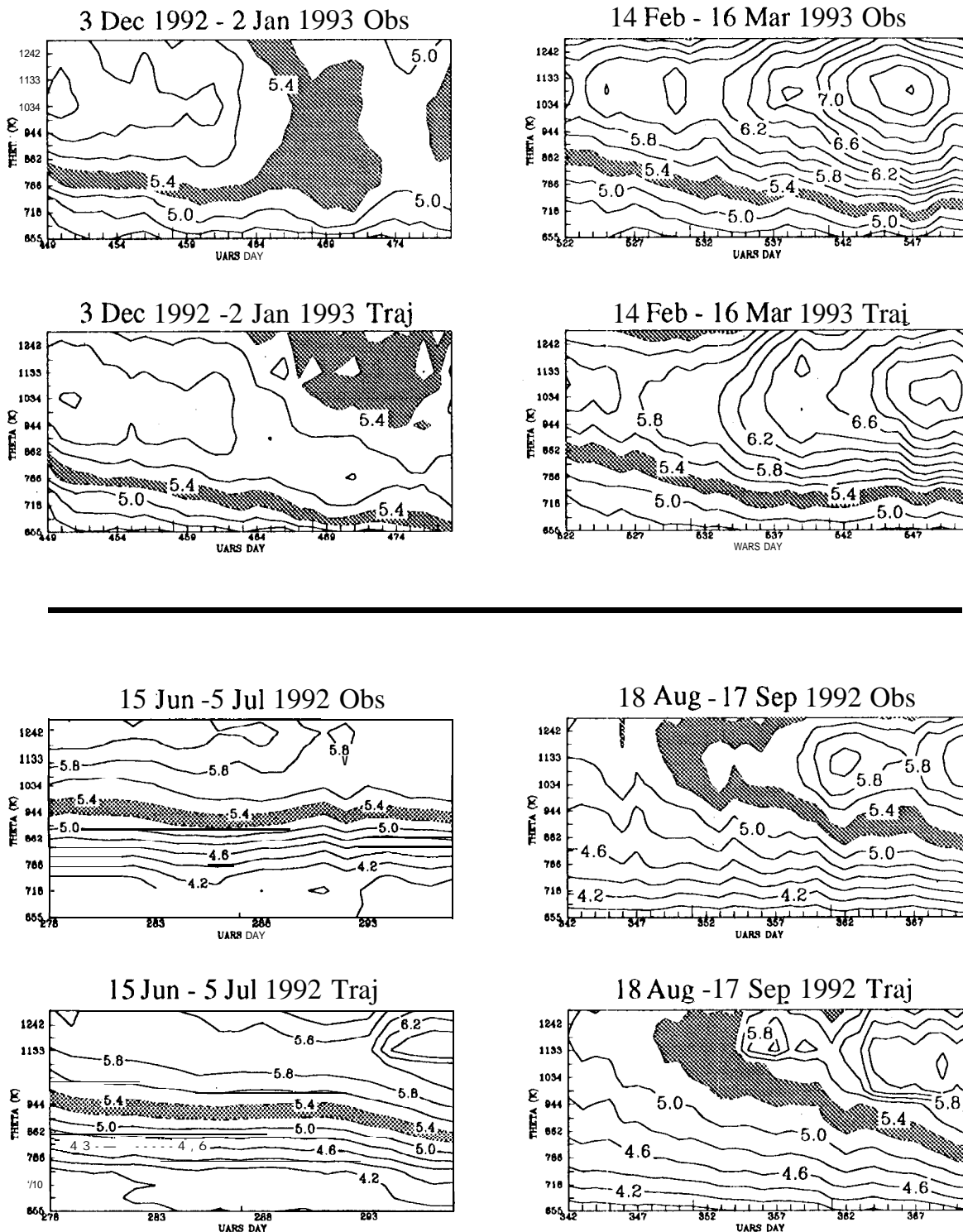


Fig. 3

465 K Ozone (ppmv)

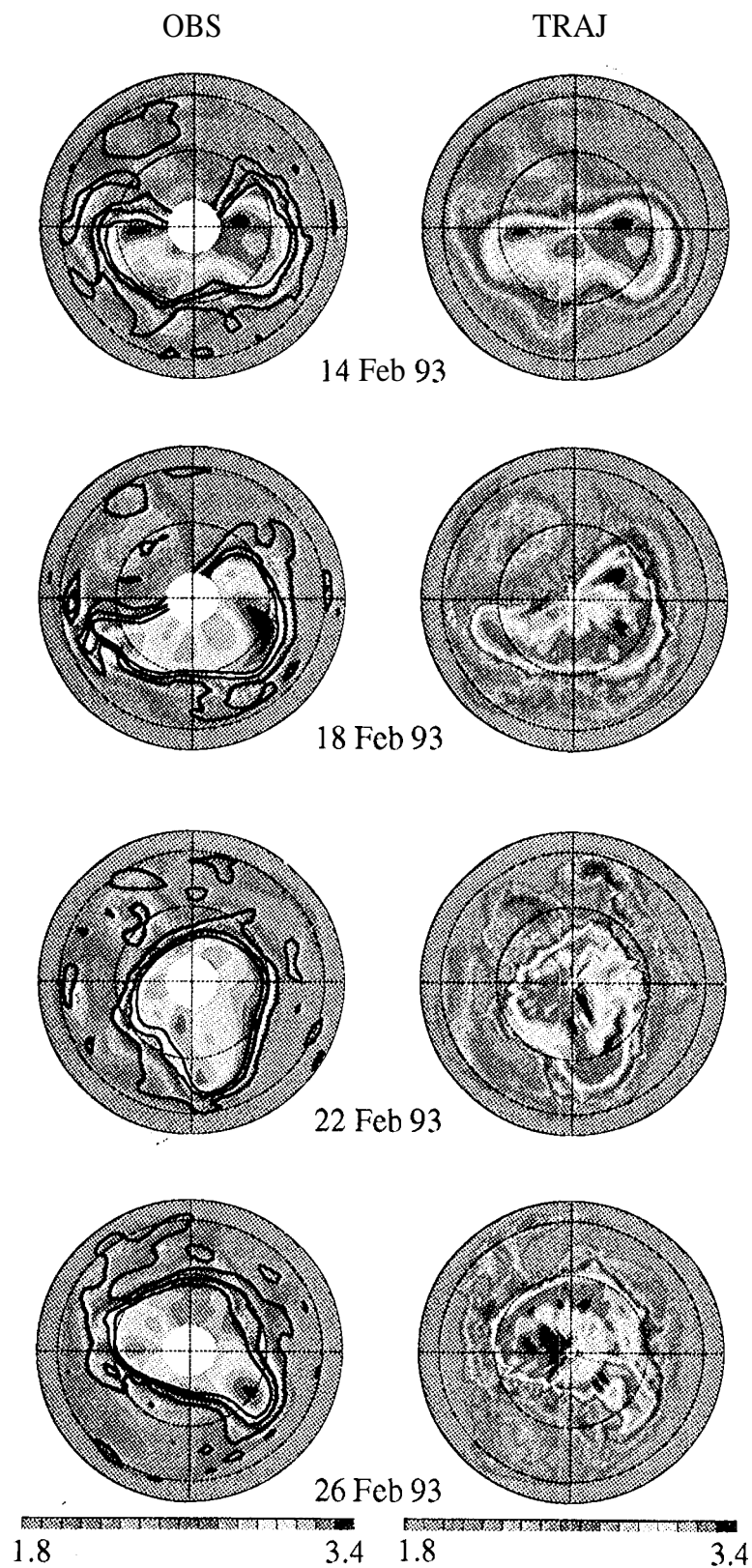
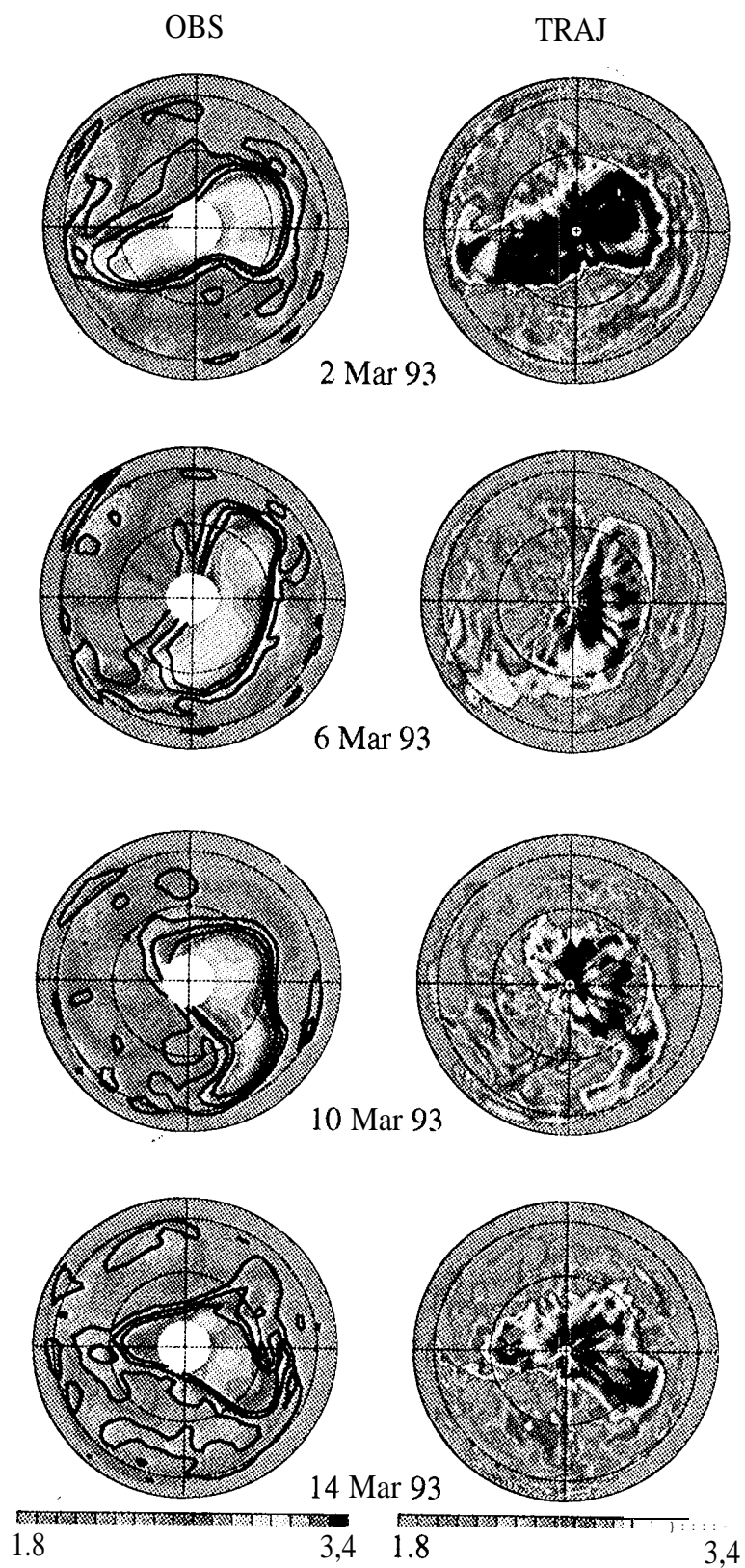


Fig. 4a

465 K Ozone (ppmv)



*Fig. 4a cont.*

465 K Ozone (ppmv)

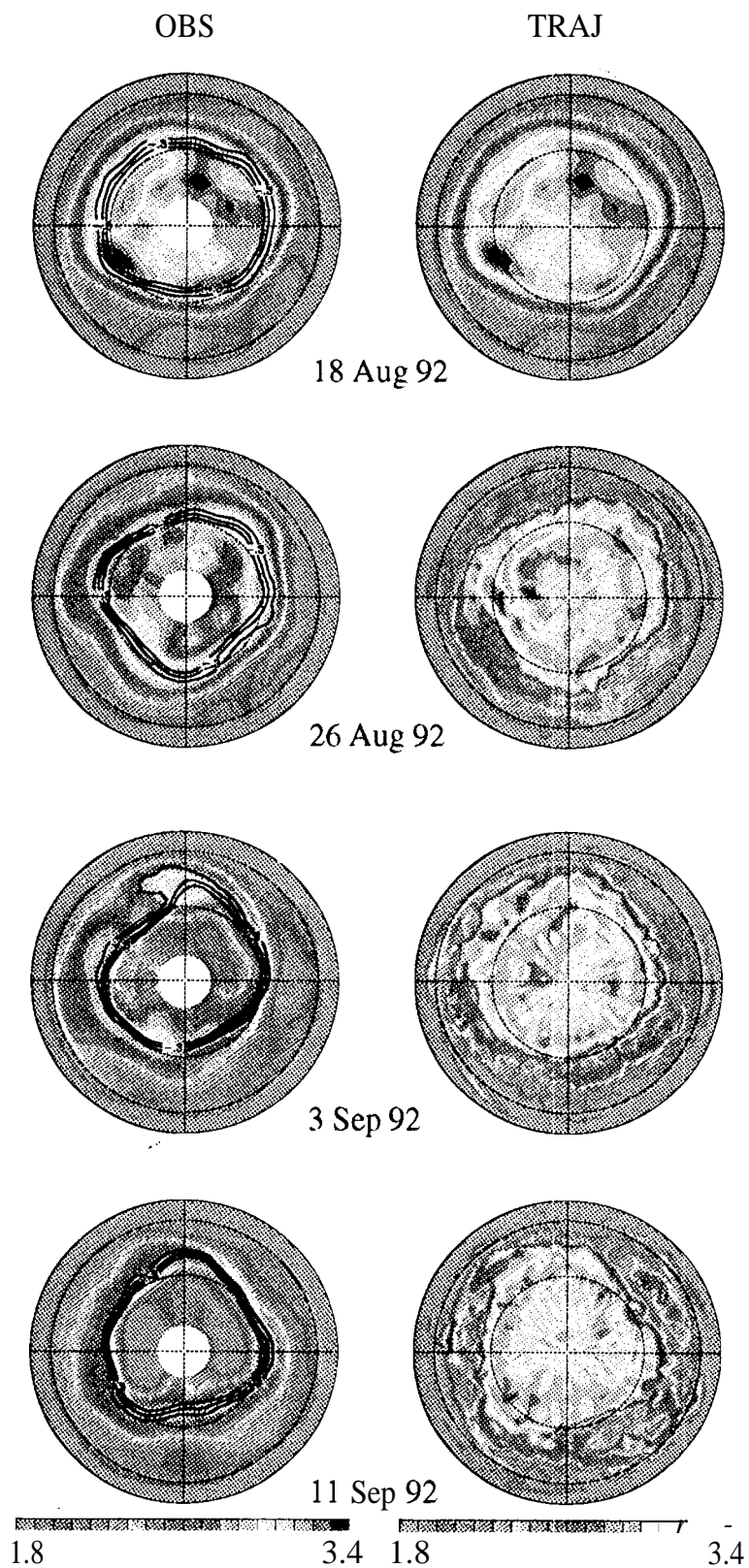


Fig. 46

# VORTEX-AVERAGED OZONE (PPMV) LOWER STRATOSPHERE

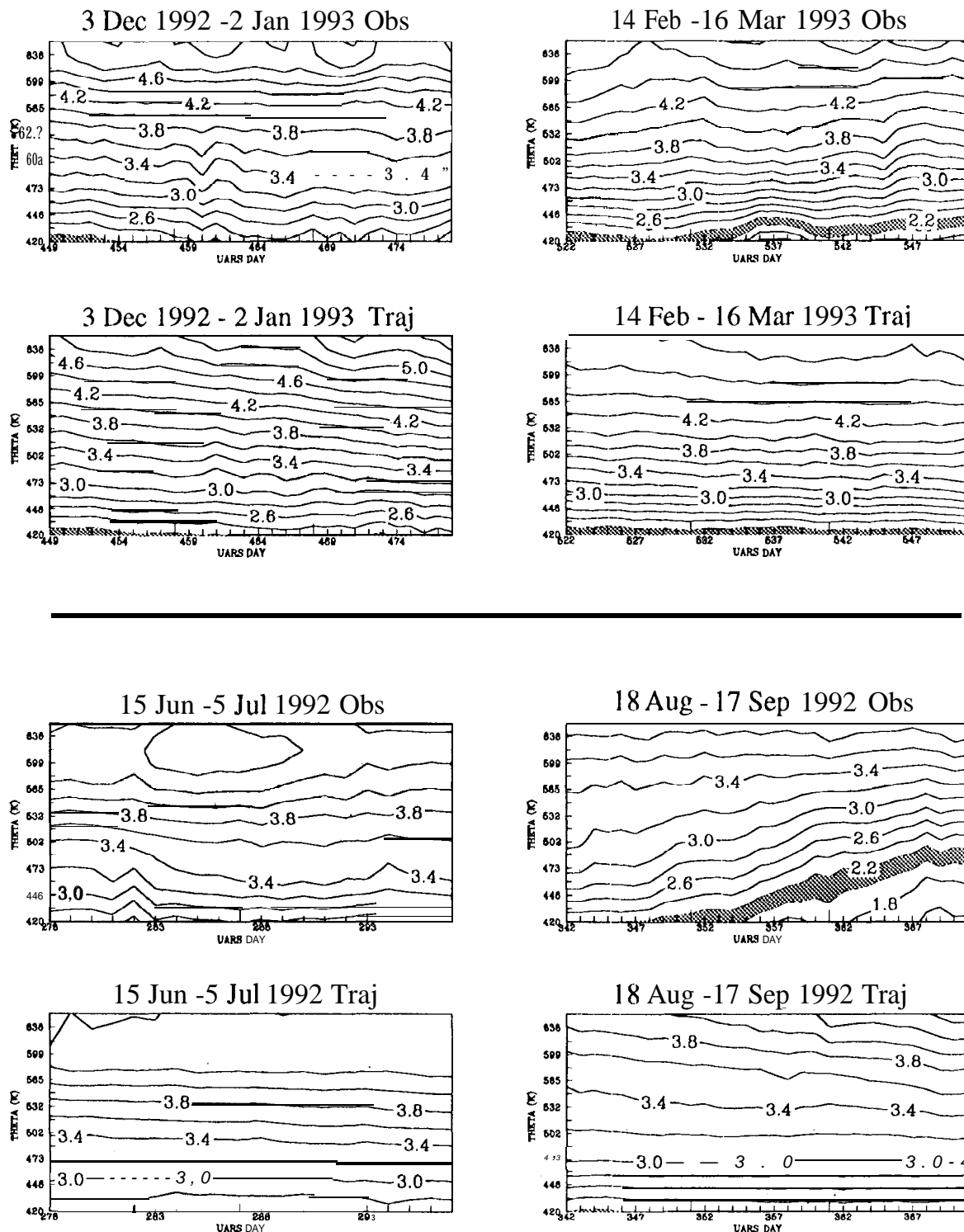


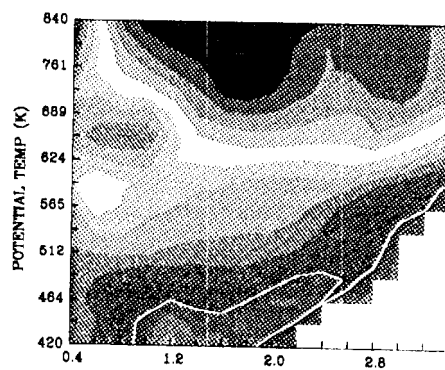
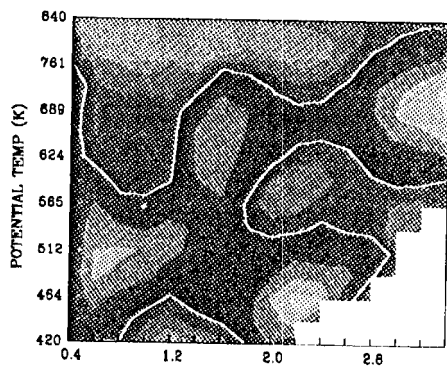
Fig. 5

OZONE CHANGE (PPMV)

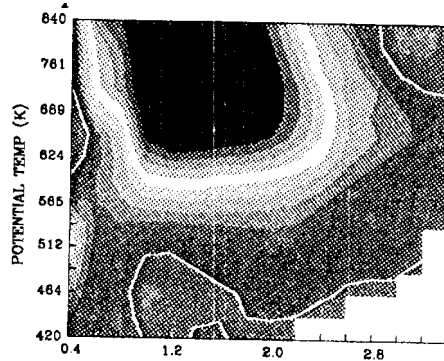
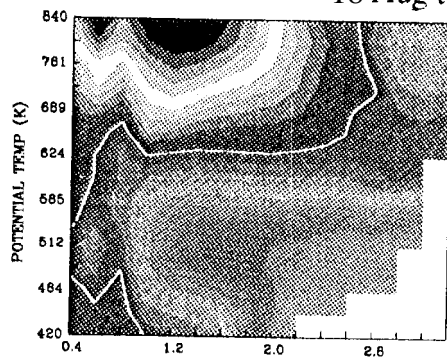
OBS

TRAJ

15 Jun to 5 Jul 1992 SH



18 Aug to 16 Sep 1992 SH



14 Feb to 15 Mar 1993 NH

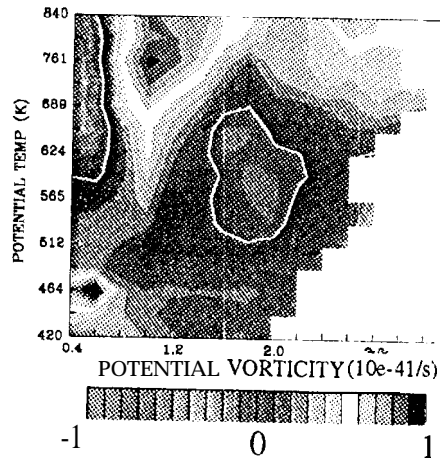
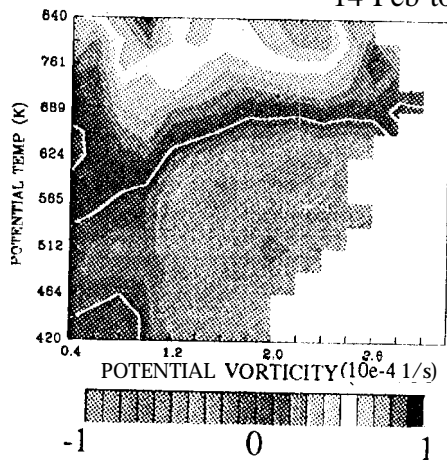


Fig. 6

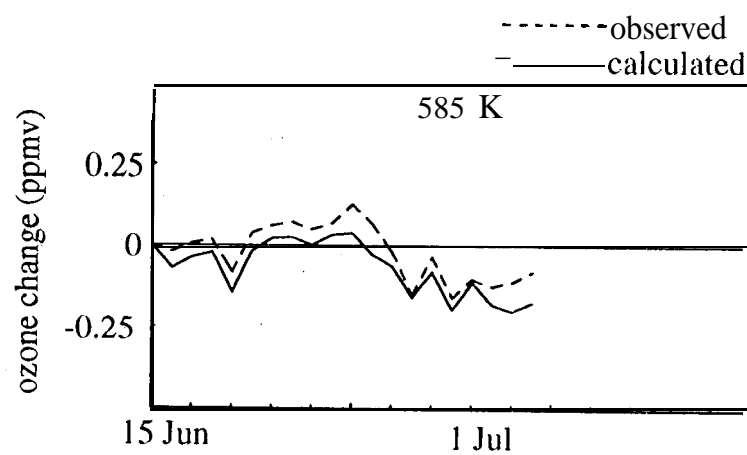


Fig. 7

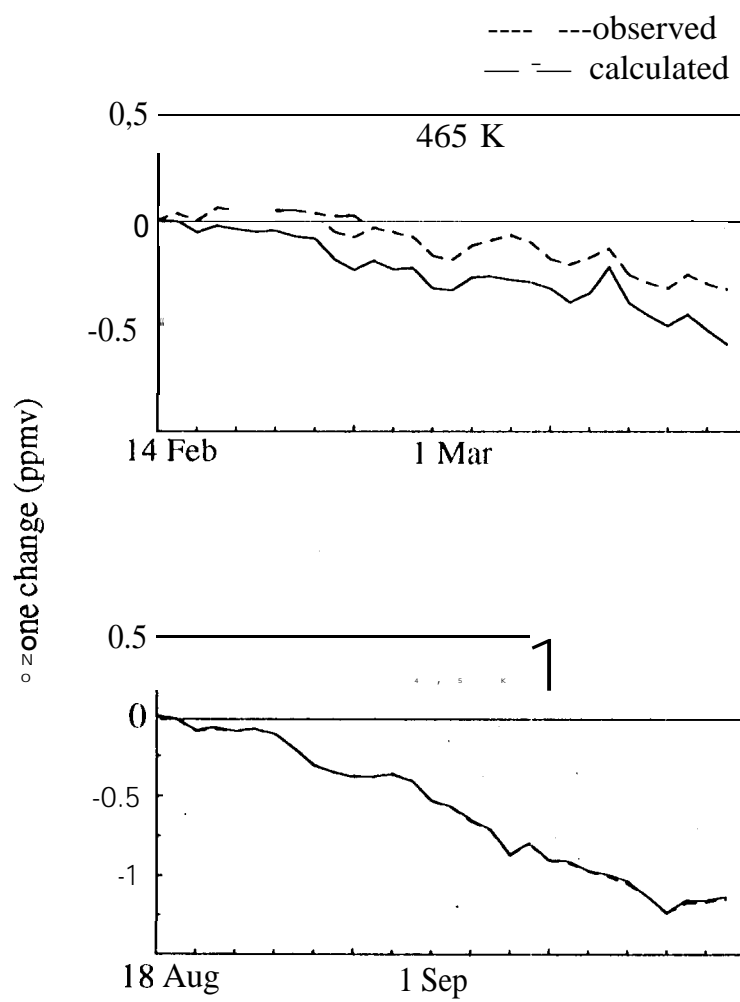


Fig. 8